Supplemental Information for:

Flexible self-supporting graphene-sulfur paper for lithium sulfur batteries

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1. Experiments

Preparation of graphene-sulfur (GS/S) paper electrode: The graphene sheets (GS) used in this work were bought from Ningbo Institute of Materials Technology & Engineering Chinese Academy of Sciences. The GS/S paper electrode was prepared as follow: 60mg of graphene sheets were first dispersed in 220 ml de-ionized water by ultrasonic treatment for 6 h, and then 1.395g $Na_2S_2O_3 \cdot 5\ H_2O$ was introduced into the graphene solution until the dissolution finished. The solution was stirred for 2 h after 20 ml 1 M HCl solution was added by drop. The GS/S paper electrode was obtained after vacuum filtration and drying.

2. Material characterization

The morphology of the as-prepared sample was observed by using Hitachi S-4800 field-emission scanning electron microscope (FE-SEM) and JSM-6700F transmission electron microscope (TEM). The content of sulfur was detected through thermogravimetric analysis by using Netzsch STA 409 PC and tested from room

temperature to **400**°C under N_2 flow with a heating rate of 5 °C min^{-1}.

3. Electrochemical analysis

The GS/S paper electrode was tested in 2025 coin cells using lithium metal foil as counter electrode and GF/A glass microfiber filter as separator. The electrolyte solution was 1M LiTFSI in a mixed solvent of tetraethyleneglycol dimethyl ether (TEGDME)/ 1, 3-dioxolane (DOL) (1:1, v/v). Each electrode has a thickness of 40-60 μm and a diameter of 14 mm. The charge-discharge performance of the cells was tested with LAND CT-2001A instrument between 1.0 and 3.0 V at 0.1 C (167.5 mA g^{-1}). The specific capacity was calculated with the mass of sulfur.

4. Figures

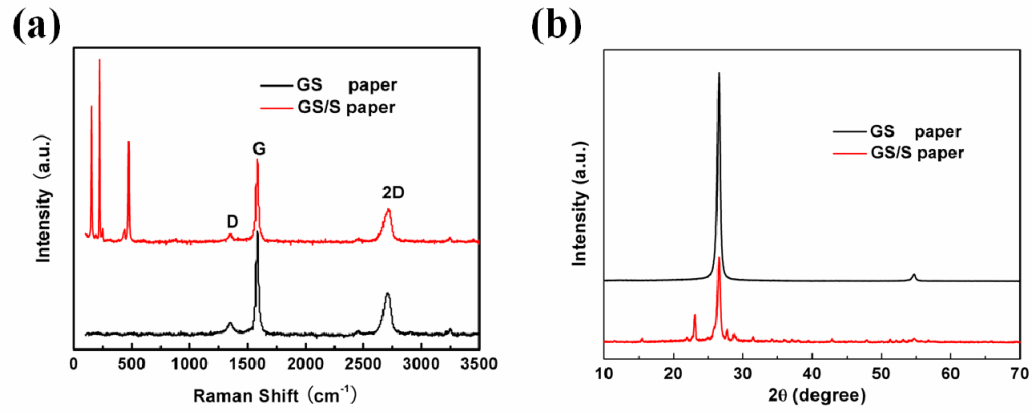

Fig S1. Raman spectras and XRD patterns of GS paper and GS-S paper electrodes. The G band at 1580 cm^{-1}, D band at 1350 cm^{-1} and the 2D band at 2697 cm^{-1} due to the GNS phase, and characteristic bands below 500 cm^{-1} due to sulfur element.

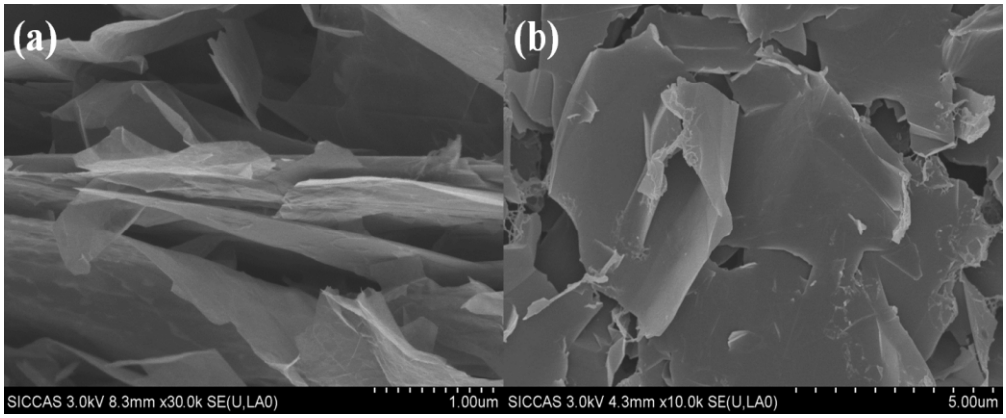

Fig.S2 Cross-section (a) and surface (b) SEM images of graphene paper

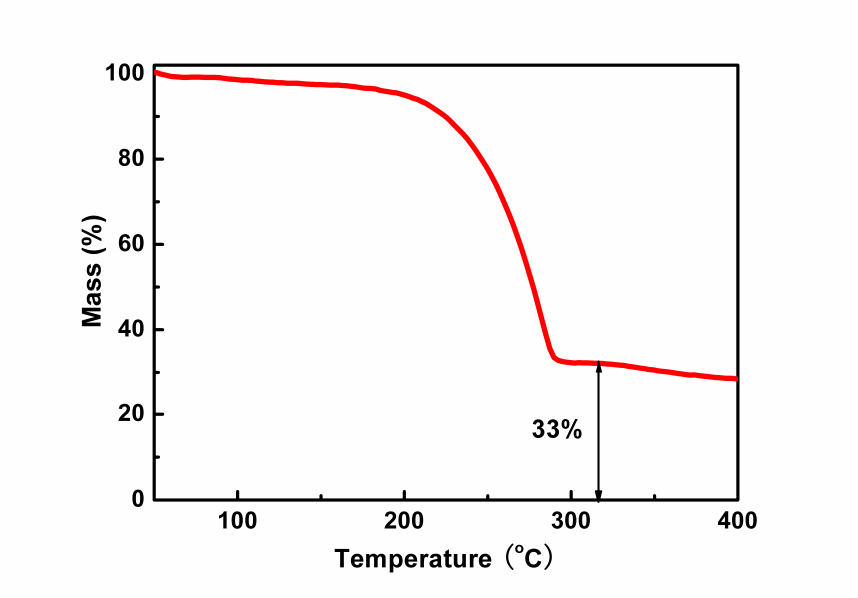

Fig S3. Thermogravimetric analysis curve of the GS/S paper electrode, showing 67wt% sulfur in GS/S paper.

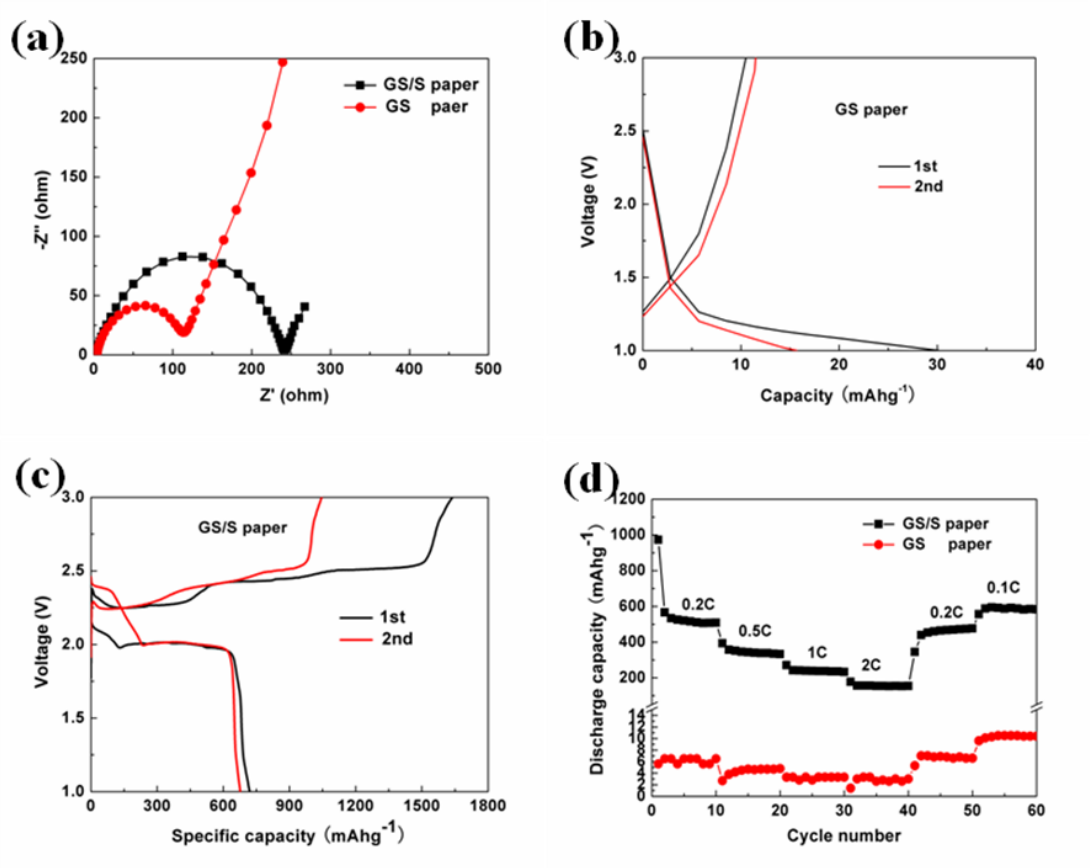

Fig.S4 (a)The AC impedance, charge-discharge curves of GS (b) and GS/S paper(c) at 0.1C (167.5mAg^{-1}) and (d) rate performance of GS paper and GS/S paper. (2C = 5.13 mA cm^{-2}).